NUCLEAR FORENSIC REFERENCE MATERIALS: SRMS 4600 & 4601 SURROGATE POST-DETONATION URBAN DEBRIS

J. L. Mann, M.A. Tyra, J.L. Molloy, R. Paul (NIST)
J. Buscaglia, J. Leggitt (FBI)
K. Pfeuffer, B. Fallon, J. Dettman (ORISE – FBI)
D. Haws, S. Castro (now AFTAC), T.D. Kelly (now AFTAC), D. Turner, J.W. McClory, (AFIT)
S. Jerome (NPL)

Goal: Deliver the first post-detonation nuclear forensic Standard Reference Materials (SRMs) to enable laboratories to establish the traceability, accuracy, and precision of nuclear forensic measurements and provide the ability to benchmark measurement methods essential to meeting the requirements of legal scrutiny.

Problem

Post-nuclear weapon detonation urban debris characterization is complicated by the high concentrations of elements like Ca, Fe, Al, and Si and other materials that would be entrained into a nuclear fireball from the local environment. This characterization can potentially identify the fuel type, weapon design, production process, and date. The resulting nuclear forensics characterization data collected must be anchored by validated measurement methods and be accurate and precise as conclusions drawn from this data will be coupled with law enforcement and intelligence information to identify who is responsible for the attack.

SRMs

Standard Reference Materials:

- Establish accuracy and precision and the traceability of nuclear forensic measurements
- allow benchmarking of investigative analytical methods meeting the requirements of legal scrutiny

NIST, as a part of its mission as the US National Metrology Institute (NMI) to deliver standards and foster homeland security, has launched the first certification campaign of measurement traceable post nuclear detonation SRMs. NIST, in cooperation with partner labs (AFIT (DoD), FBI (DOJ), and NPL) and with support from the FBI, has developed two Surrogate Post-Detonation Urban Debris (SPUD) Standard Reference Materials (SRMs) – SRM 4600 and 4601.

What is It?

SRMs 4600 and 4601 simulate the expected debris mixture from the detonation of an Improvised Nuclear Device (IND) in an urban environment. They are capable of producing fresh fission products and neutron activation products.

- The elemental composition (Table to right) of the vitrified debris materials is a composite of minerals and metals characteristic of the building materials (e.g., concrete and steel) that would be exposed to a nuclear detonation and whose resulting activation products would be found in rubble samples collected for forensics evidence.
- Post-detonation samples would also contain short-lived fission products, thus the debris materials have been doped with Uranium (U). To create the nuclear fission products, samples of the SPUD materials can be placed in a well-characterized nuclear reactor and exposed to a set neutron flux and energy, causing the encased U atoms to split and yield short-lived fission products.

	SRM 4600	SRM 4601
Element	**µg/g**	**µg/g**
Aluminium	51800	55690
Arsenic	20.3	18.7
Calcium	142000	144900
Chromium	575	500
Cobalt	45.7	46.24
Copper	240.8	262.7
Iron	195500	188400
Magnesium	3350	3160
Manganese	1812	1874
Molybdenum	263	261
Nickel	360	611
Niobium	77	78.5
Phosphorus	331	349
Potassium	2610	2718
Silicon	168800	172800
Sodium	749	734
Strontium	295	308.3
Tantalum	40.3	43.3
Thorium	16.38	16.16
Tin	29.4	28.13
Titanium	1205	1248
Tungsten	34	19.6
Uranium	62.61	81.072
Vanadium	130.8	141.3
Zinc	14	12.2
Zirconium	53.5	56.8

How it Works as SRM?

Because a known amount of U and other elements is added to the SPUD materials it is feasible to determine the concentration of the resulting fission and activation products. The subsequent irradiated SPUD glass would be chemically similar to samples that would be generated in an actual detonation, and thus can serve as a standardized reference material for nuclear forensic post detonation measurements.

Project status - Homogeneity Characterization

A priority for any reference material producer is to supply a material that is suitably homogeneous, with suitability being driven by the instrumental methods being performed by the measurement community or customer. Delivery of a homogeneous reference material is required to achieve comparability between measurement communities. Two approaches are being used to assess the homogeneity of the minor and trace elements in the SPUD materials: **microbeam X-ray fluorescence (µXRF) spectrometry** and **neutron activation analysis.** Preliminary data results are below.

Neutron Activation

Irradiation Conditions

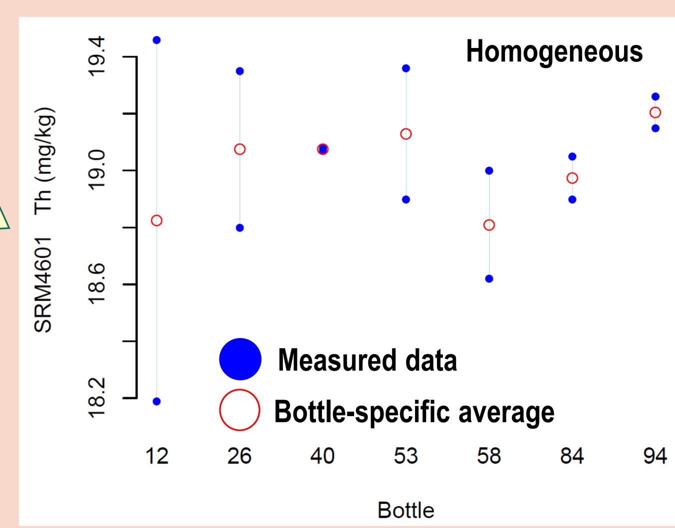
- Sample size ~ 250 mg
- Irradiated for 1 h in NIST 20 MW Neutron Beam Split-Core Reactor (NBSR)
- Neutron fluence rate of approximately ($3 \times 10^{13}\ n^{-2} \cdot s^{-1}$), thermal energies
- Irradiated at two day intervals

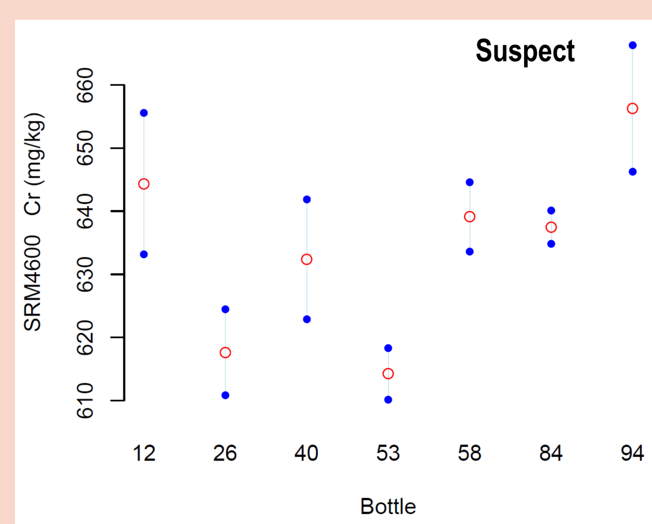
Samples that have between bottle standard deviations > the within bottle standard deviations and small p-values may be heterogeneous (e.g. Cr).

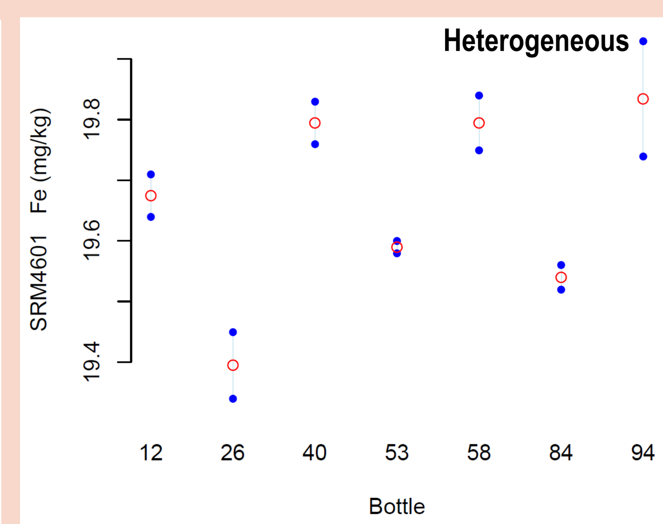
Random Effects Model

	SRM 4600 (Unat)			SRM 4601 (U235)		
Element	**τ (between bottle STD)**	**σ (within Bottle STD)**	**P-value**	**τ (between bottle STD)**	**σ (within Bottle STD)**	**P-value**
As	0.075	0.177	0.42	0.346	0.224	0.0167
Co	0.303	0.478	0.41	0.313	0.315	0.078
Cr	12.65	10.91	0.062	6.549	5.959	0.122
Fe	0.158	0.189	0.10	0.148	0.070	0.003
Mo	1.89	2.76	0.42	--	--	--
Ta	0.158	0.322	0.41	0.127	0.176	0.109
Th	0.224	0.360	0.42	0.083	0.319	0.415
W	0.089	0.476	0.42	0.109	0.410	0.419
U	0.314	0.741	0.42	1.379	1.222	0.075
U238	--	--	--	1.094	1.017	0.095
U235	--	--	--	0.300	0.227	0.030

Samples where the red circles are more scattered (in the vertical direction) than the typical distance between pairs of blue dots may be heterogeneous.







µXRF + PCA Analysis

The PCA evaluation requires:

- A 'stability run' (repeated µXRF measurements at one location on the sample) - any variance observed is related to instrument and measurement sources.
- A 'random run' (microXRF measurements are taken randomly within a defined region of interest) - any observed variability is due to instrument and measurement sources and sample heterogeneity.

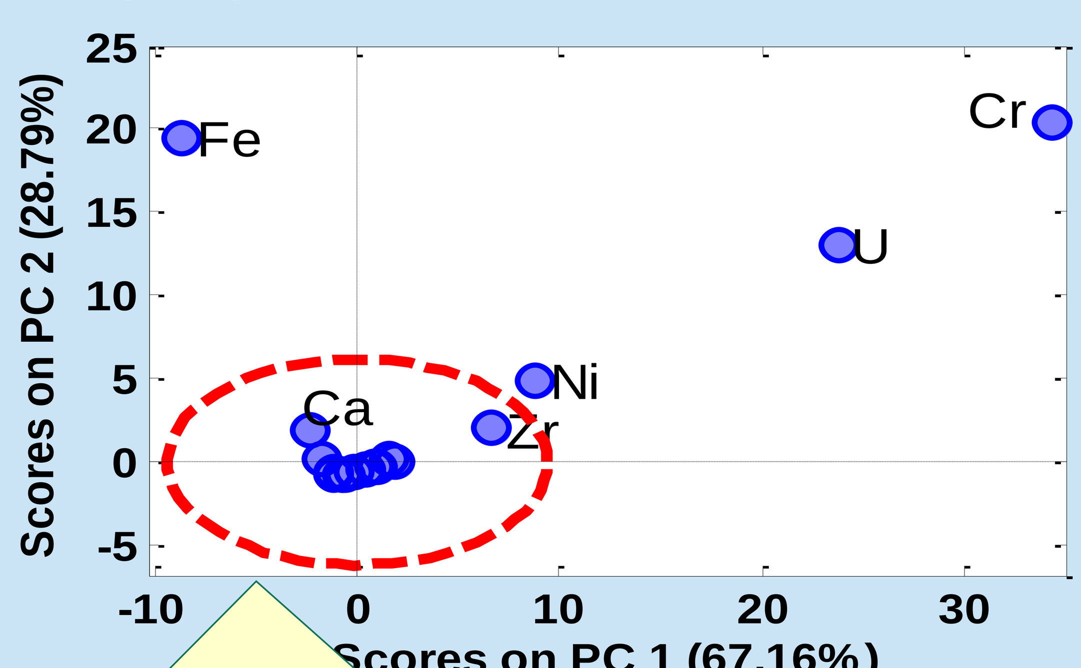


Figure 1: PCA results for one sample of SRM 4600. The red ellipse denotes three standard deviations from the center of the PCA model built on the XRF 'stability run' data obtained for this sample. The PCA model suggests that four elements (Cr, Fe, Ni and U) lie outside the ellipse and exhibit detectable "nugget effects" or sample heterogeneity**.**

The 'stability run' represents an essentially homogenous sample and can be used as a homogeneity standard to build the PCA model. The PCA model is then built upon comparison of the 'stability run' to the 'random run' to characterize variance due to sample heterogeneity as detailed in Molloy and Sieber [1, 2]. Using a combination of PCA and microXRF an estimated minimum sample mass can be derived for the entire sample where sample heterogeneity is not considered to be a large contributor to the measurement uncertainty.

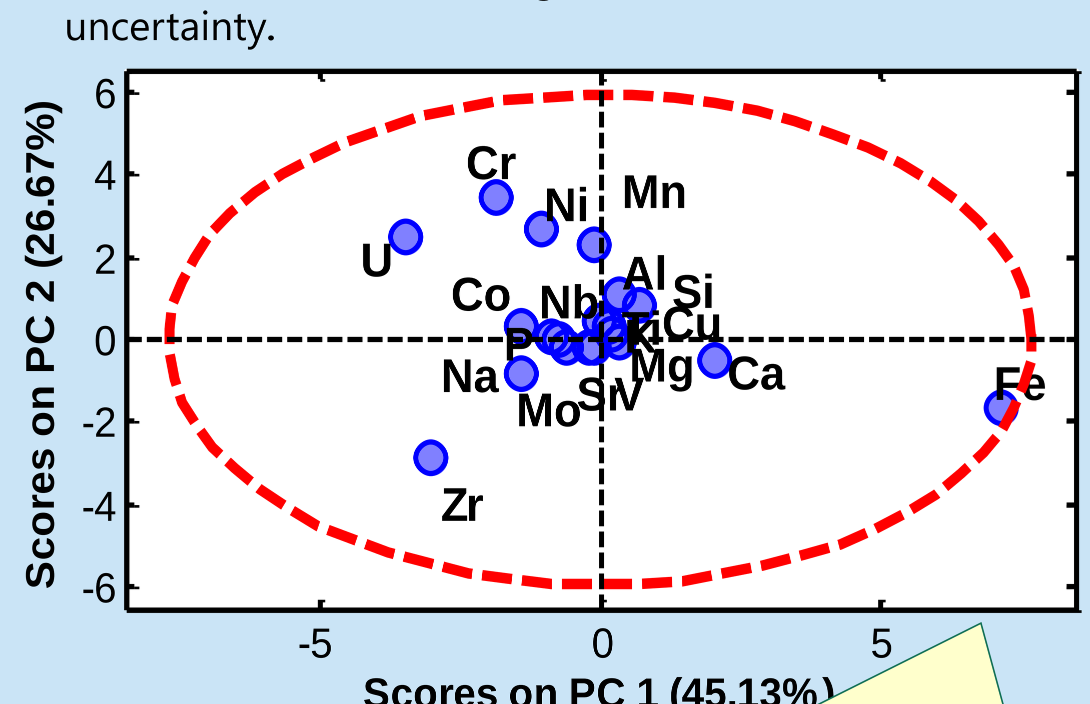


Figure 2: PCA results after averaging 250 spectra from SRM 4600. All elements fall within the red confidence ellipse indicating no measurable nugget effects at a sample size of 1 mg.

Takeaways

- Elements measured by INAA are less heterogeneous in SRM 4600 then in SRM 4601 (for 250 mg sample).
- Only Cr seems to be heterogeneous for SRM 4600 for 250 mg sample.
- As, Fe, and U are suspect for SRM 4601; Co is suspicious at 250 mg.
- Preliminary µXRF + PCA analysis of SRM 4600 suggests that Cr, Fe, Ni, and U maybe heterogeneous at 50 mg sample size.
- Data suggest that Cr is heterogeneous for SRM 4600 at any sample size and that for smaller sample sizes (< 250 mg) Fe, Ni and U may also be heterogeneous.
- HOWEVER, thus far only one sample has been run through PCA analysis so the suggestion that only 1 mg is required for all the elements to be homogeneous should be taken lightly.

Contact Jacqueline Mann
NIST, 100 Bureau Drive, Stop 8462, Gaithersburg, MD 20899, USA

phone: 301-975-4472, email: jmann@nist.gov

1. J. MOLLOY and J. SIEBER, "Classification of microheterogeneity in solid samples using mu XRF," *Anal. Bioanal. Chem.*, 392, 995 (2008).
2. J. MOLLOY and J. SIEBER, "Assessing microscale heterogeneity of batches of reference materials using microbeam XRF," *X-ray Spectrom.*, 40, 306 (2011).